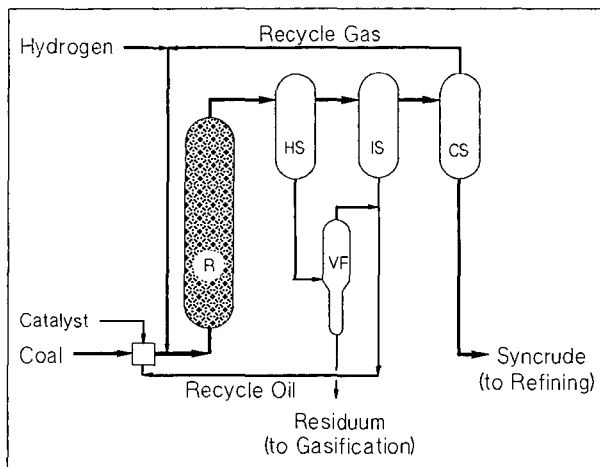


**Figure 1. DT coal hydrogenation process, simplified flowsheet.**  
*R, reactor. HS, IS, CS, hot-, intermediate-, cold separator.*  
*VF, vacuum flash unit*



**Figure 2. ASTM D-2887 boiling analyses of product and recycle oil from IGOR<sup>+</sup> coal hydrogenation (dotted lines) and DT coal hydrogenation (solid lines).**

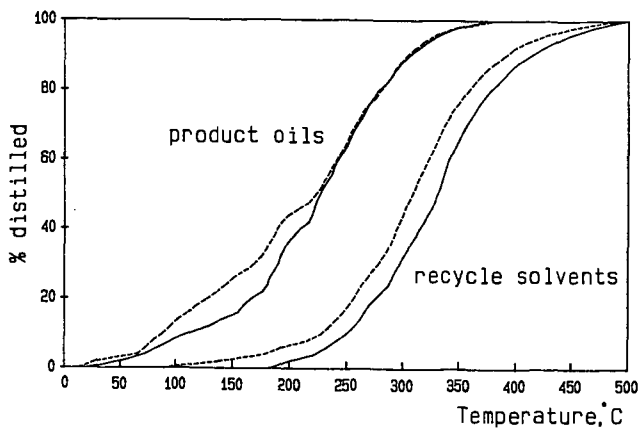


Figure 3. IGOR process, simplified flowsheet.

FB, fixed-bed catalyst. Other symbols, see Fig. 1.

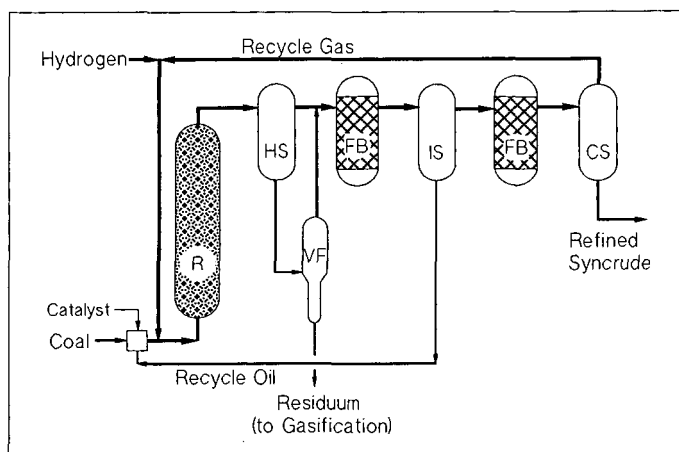
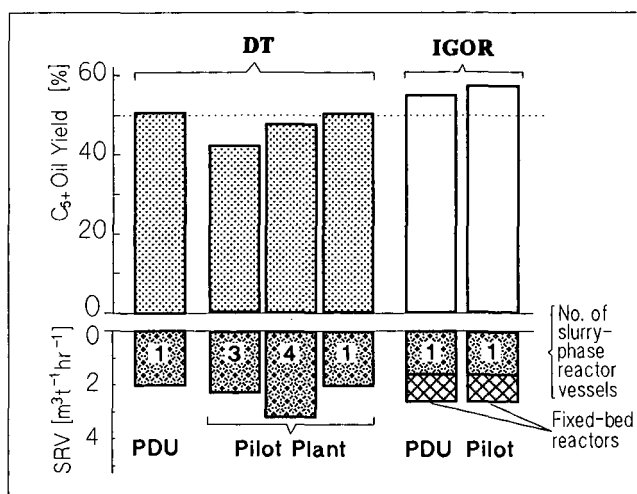


Figure 4. DT and IGOR process scale-up history.

SRV, specific reactor volume, d.a.f. coal basis.



## **IGOR - TAKING THE SHORT CUT IN COAL HYDROGENATION**

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**Keywords:** coal hydrogenation, upgrading, integrated refining, IGOR process

### **ABSTRACT**

The primary liquids obtained from conventional coal hydroconversion processes all require extensive downstream hydrotreating. With a hydrotreating reactor appropriately integrated into a coal hydrogenation scheme, reformer/hydrocracker feed quality specifications can directly be attained, with only one process unit. A further modified arrangement even leads to significantly increased hydrocarbon oil yields from coal. The developing of the novel process, integrated gross oil refining (IGOR), is described. Results and experiences from PDU and from pilot plant operation are discussed.

### **INTRODUCTION**

Several coal hydrogenation processes have been developed to technical maturity since coal-to-oil technologies were given a chance again. A good number of novel features have been successfully incorporated, but the competing schemes can all still be seen as mere modifications of the conventional high-pressure hydrogenation process: Apart from the processing steps they have all in common (slurry preparation, solvent recycle, indirect pre-heating, asphaltenes & solids separation, etc.), there is another joint feature. The primary coal-derived distillates ("syncrudes") are unacceptably high in oxygen and nitrogen contents and require extensive further refining prior to hydrocracking, reforming, or use as a marketable fuel whatsoever. (e.g.<sup>1</sup>) Particularly in view of the black sediments that form even during short-term storage, and which are severely poisonous to upgrading catalysts, these syncrudes should only be taken with some reservations as the clean distillates they seem to be.

A further aspect which requires attention is, that the raw syncrudes have biological implications and are perceived as a potential health hazard at least in the working field. In syncrudes, primarily polynuclear aromatics, aromatic nitrogen bases, benzene, and phenols as well are of relevance to health considerations. It has therefore been recommended that the available coal hydrogenation processes be operated or modified such as to yield lower boiling and preferably saturated coal oils.<sup>2</sup> Developing an entirely different hydrogenation scheme might be another option, yet some way of integration of refining and saturation should already help to improve the technical, environmental, and probably also the economical preconditions for a future coal hydrogenation industry.

### **EXPERIMENTS AND RESULTS**

A 0.2 t/d PDU and a 200 t/d pilot plant have been operated within the development work presented in this paper. All runs referred to in the following were carried out under 30 MPa total pressure, with a German HVBA feed coal and red mud as the catalyst in coal conversion. Commercial Ni-Mo-alumina catalysts were used in the fixed-bed reactors.

As for PDU operation, normally one week runs with the conditions maintained unchanged made a good basis for reliable data. With any mode of integrated refining, the runs were typically extended to last one month or more, primarily in order to make possible catalyst deactivation visible. For the same reason, these runs were repeated after several months.

### *DT Process Operation.*

In order to explain the procedures and equipment used, we will firstly refer to the - earlier - DT process configuration (*Figure 1*), as this process provided the basis for the further developments.

A coal slurry is prepared from dried, ground coal, some powdered once-through catalyst, and a distillate-type process recycle oil. After treat gas addition (high-pressure recycle gas plus hydrogen make-up), the slurry is passed through heat exchangers and pre-heated to 420 °C. The upflow tubular reactor is operated as a rule at 475 °C. Still under process pressure, the reaction products are separated with a series of coolers/heat exchangers and stripper volumes: At about 450 °C minerals, unconverted coal, and little- and nonvolatile organic liquids are removed from the vapors and gases and let down from the hot separator. In a vacuum flash unit, some high-boiling overhead oil is recovered from the hot slurry, and a hot liquid underflow that contains around 50 % solids. The overhead oil is used as a recycle oil component, the underflow residuum as a liquid gasifier feed for hydrogen production.

The oil fraction condensed through appropriate temperature reduction (to around 300 °C) of the hot separator overhead vapors, is obtained as the intermediate separator letdown in just the amount required for coal slurry preparation. By further cooling of the high-pressure vapors and gases, the net syncrude yield condenses, along with process water which separates readily from the organic phase. - The high-pressure gases that leave the cold separator are oil-scrubbed for hydrocarbon gas reduction, and recycled to the hydrogenation unit.

### *DT Process Modification: Integrated Net Oil Refining.*

Much as with practically any proven coal hydrogenation process, the syncrudes recovered from DT coal conversion will require extensive refining. With respect to their eventual utilization as motor fuels and possibly jet fuels, DT syncrudes have favorable boiling ranges (end points lie around 330 °C). But in order to meet the existing specifications for marketable fuels and hydro-conversion feed oils, respectively, the high concentrations of oxygen and nitrogen containing compounds as well as fused-ring aromatics need be radically reduced. Because of the essentially cyclic nature of coal-derived oils, comparatively severe conditions are required in the hydro-refining of such feed.

We found that it is not only viable but even advantageous to integrate a hydrotreating and saturation stage into a coal hydrogenation plant rather than running the units separately.<sup>3</sup> With a fixed-bed catalytic reactor arranged downstream the intermediate separator, the product oil vapors are hydrotreated still before they are condensed and let down. Thus, by taking advantage of both the high-pressure, hydrogen-rich gas as well as the sensible heat of the separator overhead stream, appreciable savings in investment and energy costs can be realized.

Extended PDU runs established that the product oil qualities obtained with separate refining are attained with integrated refining as well. The total heteroatoms concentration - nitrogen, oxygen, and sulfur - can be kept well below 10 mg/kg. Oil yields from separate and from integrated refining, respectively, are naturally about the same: With the little reactive German bituminous feed coal, the yields of refined C<sub>4+</sub> oil amount to 54 % w/w on dry, ash-free coal in both cases.

### *Integrated Gross Oil Refining.*

Further coal hydrogenation tests included a modified integrated refining scheme in which the fixed-bed reactor was arranged such as to hydrotreat not only the net product but also the oil continually recycled for coal slurry preparation ("gross oil refining"). The heavy distillate fraction - vacuum flash overhead oil - was injected before the reactor. With a liquid hourly space velocity four times the previous one and with the feed boiling range much higher at that, several hundred mg/kg hetero atoms were left in the net product oil. Net oil yield, however, had surprisingly soared to more than 60 %. Tracer and other investigations have made clear that the yield improvement is brought about thanks to a much extended liquid/solid residence time in the coal conversion reactor.<sup>4</sup> The "driving force" is enhanced oil stripping which in turn is due to the more readily volatile recycle oil, see the boiling curves plotted in *Figure 2*.

With nitrogen contents between 80 and 100 mg/kg and even higher oxygen levels, the product oil fails to meet minimum feed specifications for further processing over bifunctional catalysts. In other words, the extra oil yield is obtained at the expense of quality, which additional hydrotreatment indispensable.

The process recycle oil, once equilibrated through continual recycling, will - simply speaking - behave as an inert diluent that does not undergo further hydrogenation reactions. Seen from this point of view, the fixed-bed reactor is merely treating the net product oil, though at an unfavorably high space velocity. Therefore, it seemed promising to apportion the catalyst filling into two separate but smaller reactor volumes, with the first one employed for gross-oil- and the second one for net-oil-treatment only (see *Figure 3*).<sup>5</sup> The results received with this mode of operation fully answered the expectations, as both high oil yield and perfect quality were obtained now at the same time. This process mode was named IGOR<sup>+</sup>. It compares favorably with other processes as can be concluded from *Table 1* - in particular when considering the rather low reactivity of German bituminous coal. A synopsis of PDU results obtained with the various process modes discussed above is shown in *Table 2*.

## DISCUSSION

### *Process operation experiences.*

Even if quite obvious, it should be noted that IGOR<sup>+</sup> product oil, thoroughly different from any syncrude, is water-white, stable in storage, and completely free of "that certain coal-oil smell". Less obvious, most of the process internal oil streams, particularly the recycle solvent inventory, have a merely faint, "clean" odor and separate easily from water. Moreover thanks to IGOR<sup>+</sup>, any biotoxic potential suspected in the previous process liquids has in all probability been removed. The improvement of the environmental conditions, in particular at the PDU with its frequently opened containers and its occasional leaks and spills, is unmistakable.

With the IGOR modes of operation, a new situation was encountered in coal slurry handling: Density and viscosity of the recycle oil had become too low to keep the solids suspended so that settling, pump failures, and other problems emerged. As a countermeasure, the earlier slurry viscosity was re-adjusted to its previous value by simply raising solids concentration from 42 to 51 % (coal-to-oil ratios, 1:1.4 and 1:1, resp.). As the slurry feed rate was not correspondingly reduced, this meant an increase in specific coal feed rate by 20 %. It turned out that, different from the DT process, oil yield suffered only little from the elevated specific coal feed rate. At the same time, apparently due to the reduced liquid feed load to the first fixed-bed reactor, product analyses showed no sign of oil quality reduction. Thus, a slurry solids concentration of more than 50 %, and a specific feed rate of 0.6 kg m.a.f coal per hour and per liter of reactor volume, became features of IGOR "standard" operation conditions.

Coal conversion temperatures in the IGOR process are somewhat lower than in the DT process which results in a reduced gas make and thus, in a comparatively lower hydrogen demand. Some extra hydrogen, on the other hand, is consumed in the methanation of the carbon oxides which takes inevitably place at the highly active refining catalysts. The coals that are typically fed to the process form only little carbon oxides (roughly 1 % on coal under DT process conditions) so this is certainly not too great a drawback. But a great advantage was found as well:

In any conventional coal hydrogenation process, continuous purge water injection is mandatory in order to prevent plugging of high-pressure gas lines with carbon dioxide derived salts, essentially ammonium carbamates. Hence sour water output is much higher than would be brought about from only hydrodeoxygenation and the coal moisture. With integrated refining there are no carbon oxides left, so purge water is no longer necessary. At the same time, refining has completely removed the phenols. Accordingly there is not only much less waste water to be treated but it does also no longer contain phenols that had to be dealt with. Total Organic Carbon had averaged 10,000 mg/kg in the conventional process (DT); with IGOR<sup>+</sup>, TOC values never exceeded 50 mg/kg.

### Scale-up Experiences and Results.<sup>7</sup>

In most if not all direct coal liquefaction projects that were continued to pilot scale, the syncrude yields obtained in the PDUs could not be reproduced on a hundred-tons-per-day level [LIT X]. Initially, this was also true for the DT process. A single volume (11 l) made the PDU coal conversion reactor while a tanks-in-series reactor system was used on pilot plant scale, much as with the I.G. technology development. At identical specific coal feed rates, three tubular reactors (5 m<sup>3</sup> each) gave significantly less oil yield on coal than had been obtained with the PDU. Even with a much lower specific feed rate - a fourth 5 m<sup>3</sup> cylinder added - and with the reaction temperature elevated by 10 K in addition, oil yield still remained slightly below the PDU results. Contrary to all expectations, this changed when the cascade was replaced by a 15-m<sup>3</sup> capacity, cheaper-to-fabricate "large-volume reactor". Now the results were exactly the same as those in the 1,400 times smaller unit.

The process was tested on pilot scale directly with the large-volume reactor; as the fixed-bed catalytic reactors, the 5-m<sup>3</sup> cylinders were used. The results obtained were even somewhat more favorable than in the small-scale tests. In Figure 4, the above development history is illustrated and reviewed.

With the large KOHLEÖL pilot plant at Bottrop, still another benefit of the IGOR process could be demonstrated. It has been possible to bring the coal slurry up to reaction starting temperature exclusively through heat exchange with reaction product streams and preheating of the treat gas. In other words the slurry preheater, always an expensive and delicate component of the plant, was no longer necessary.

### CONCLUSION

More recently in the PDU, a different bituminous coal - much more reactive yet higher in minerals at the same time (11 % ash, dry) - was processed under "standard" IGOR+ operation conditions. In the product oil, a total hetero atoms content of less than 5 mg/kg was achieved, and the C<sub>4</sub>+ yield came as high as 66 % on d.a.f.coal.

Thus far in IGOR process testing, with totalized operation periods of up to 330 days, in neither of the two fixed-bed reactors were observed signs of serious catalyst deactivation. Presently, the only limitation foreseen is with feed coals that have too high an oxygen content and thus excessive carbon oxides formation, so that uneconomically much hydrogen might be spent in methanation.

The most apparent advantages of the process over its predecessors can be summarized as follows. IGOR+ means:

#### Reduced

- equipment needs
- recycle oil flow
- hydrocarbon gas formation
- waste water treatment requirements

#### Eliminated

- purge water injection
- carbon oxides
- phenols
- polynuclear aromatics & aromatic amines
- slurry preheater

#### Increased

- oil from coal yields
- specific coal feed rate
- coal concentration in the slurry feed

#### Improved

- product quality
  - naphtha cut ready for reforming,
  - gas oil cut ready for hydrocracking
  - or blending with diesel fuel
- working place environment
- overall plant operation
- process efficiency and economy

Rather than by splitting apart coal-to-oil conversion into discrete stages that can be individually optimized but will also suffer from individual idiosyncrasies, good yields of clean hydrocarbon liquids are also accessible in a more straightforward method using a relatively simple, integrated hydrogenation process.

## ACKNOWLEDGMENT

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Table 1. Syncrude yields and properties from different coal hydrogenation processes.  
[Data partly after Peluso et al.<sup>6</sup>]

PROCESS:		EDS	H-Coal	ITSL	SRC II	DT	IGOR <sup>+</sup>
Feed coal		Illinois # 6		Indiana # 5	Kentucky # 9	Ruhr Gasflamm, Prosper	
- Carbon	% d.a.f.	77	78	78	79.5	85	
C <sub>4+</sub> Oil yield	% on d.a.f. coal	39	50	56	43	55	60
Oil Inspection:							
- Boiling end	ASTM D86, °C	520	520	450	480	330	331
- Hydrogen	%	11.2	10.7	10.7	9.6	10.0	13.7
- Oxygen	mg/kg	15,000	15,000	7,000	35,000	27,000	< 5
- Nitrogen	mg/kg	3,000	3,000	4,000	1,400	9,000	< 2
- Sulfur	mg/kg	800	800	800	2,000	800	< 1

Table 2. Feed and product characterization for DT and IGOR process modes.

PROCESS:		DT	IGOR	IGOR <sup>+</sup>
Feed Coal		Ruhr Gasflamm, Prosper (HVBA)		
Proximate analysis				
Ash			5.1	
Volatile matter	% dry		36.5	
Elemental analysis				
Carbon	% d.a.f.		84.8	
Hydrogen	"		5.6	
Oxygen	"		7.2	
Nitrogen	"		1.5	
Sulfur	% dry		1.0	
PDU Operation Conditions:				
Specif. d.a.f. coal feed rate	kg ltr <sup>-1</sup> hr <sup>-1</sup>	0.5	0.5	0.6
Coal/oil ratio in slurry feed	kg kg <sup>-1</sup>	1 : 1.4	1 : 1	1 : 1
Total pressure	MPa	30	30	30
Reaction temperature	°C	475	470	470
Catalyst in coal conversion	(dispersed)	red mud	red mud	red mud
Catalyst in hydrotreating	(fixed bed)	—	Ni—Mo	Ni—Mo
Hydrogen consumption	% on d.a.f. coal	6.7	9.3	9.0
Product Yields:				
C <sub>4+</sub> Net product oil	% on d.a.f. coal	55	61	60.5
Flash residuum [a.f. basis]	"	24	20	21
C <sub>1</sub> —C <sub>3</sub> Hydrocarbon gases	"	20	18	17
Reaction water & inorg. gases	"	7.5	10	10
C <sub>5+</sub> Oil Inspection:				
Density @ 20°C	g cm <sup>-3</sup>	0.930	0.844	0.833
Boiling analysis, ASTM D86				
I.B.P.	°C	96	87	80
10 %	"	144	127	115
30 %	"	196	183	175
50 %	"	222	226	220
70 %	"	250	252	251
90 %	"	290	289	289
F.B.P.	"	330	331	331
Elements				
Hydrogen	%	10.0	13.0	13.7
Oxygen	mg/kg	9,000	85	< 5
Nitrogen	mg/kg	27,000	160	< 2